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(54) Detergent gel composition

(57) Stable aqueous detergent gels wholly or predominantly in hexagonal liquid crystal form comprise a fatty acid methyl ester sulphonate and/or an alpha-sulpho fatty acid salt, an optional auxiliary surfactant (soap, or a primary non-soap anionic surfactant such as a primary alkyl sulphate), an optional additive such as urea, and water. Alpha-sulpho fatty acid salts, if used alone, are present wholly or predominantly in substituted ammonium salt form, or are other than sodium salts.

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GB 2 179 055 A

SPECIFICATION

Detergent compositions

5 TECHNICAL FIELD

The present invention relates to detergent compositions in the form of a stable transparent, translucent or opaque water-soluble gel which is wholly or predominantly in hexagonal liquid crystal form. The compositions of the invention are especially suitable for washing dishes or other hard surfaces, but are also of use for other cleaning purposes, for example, fabric washing.

BACKGROUND AND PRIOR ART

US 2 580 713 (Wood/Procter & Gamble) describes paste or gel detergent compositions, in hexagonal or "middle" phase, based on certain anionic surfactants, notably triethanolamine lauryl sulphate and triethanolamine lauryl sulphate/soap mixtures.

GB 2 155 031A (Unilever) published on 18 September 1985, describes and claims detergent compositions in hexagonal phase gel form, containing alkylbenzene sulphonate or dialkyl sulphosuccinate, urea and water.

Turkish Patent No. 21612 (Mintax), laid open to public inspection on 5 December 1984, also discloses detergent gels containing alkylbenzene sulphonate, urea and water, but contains no reference to hexagonal phase.

We have now discovered that stable hexagonal phase gels can be based on fatty acid methyl ester sulphonates and/or alpha-sulpho fatty acid salts, provided that if only the latter material is present it is at least partially in the form of the salt of a substituted ammonium cation.

DEFINITION OF THE INVENTION

The present invention accordingly provides an aqueous detergent composition comprising a gel wholly or predominantly in hexagonal liquid crystal form, wherein the gel comprises:

(a) from 5 to 95% by weight of a fatty acid methyl ester sulphonate and/or an alpha-sulpho fatty acid salt,

(b) optionally from 1 to 75% by weight of an auxiliary surfactant selected from fatty acid soaps; non-ethoxylated micelle-forming non-soap surfactants having an anionic head group and an aliphatic or araliphatic or araliphatic hydrocarbon chain containing from 10 to 20 aliphatic carbon atoms, the anionic head group being positioned terminally or next to terminally in the hydrocarbon chain, and mixtures thereof; the total amount of components (a) and (b) being from 25 to 95% by weight,

(c) optionally 1 to 45% by weight of an additive which is an anionic or nonionic water-soluble non-micelle-forming or weakly micelle-forming material having a polar head group and optionally an aliphatic or araliphatic hydrocarbon chain containing at most 6 carbon atoms, and

(d) water,

with the proviso that if component (a) consists wholly of an alpha-sulpho fatty acid salt, the alpha-sulpho fatty acid is wholly or predominantly in substituted ammonium salt form.

DISCLAIMER

Our copending application No. 8619788 of even date, claiming the priority of British Patent Application No. 85 20548 filed on 16 August 1985, describes and claims an aqueous detergent composition comprising a gel wholly or predominantly in hexagonal liquid crystal form, wherein the gel comprises:

(a) from 5 to 85% by weight of a non-ethoxylated micelle-forming non-soap surfactant having an anionic head group and an aliphatic or araliphatic hydrocarbon chain containing from 10 to 20 aliphatic carbon atoms, the anionic head group being positioned terminally or next to terminally in the hydrocarbon chain, and

(b) a second component comprising

(b)(i) from 1 to 75% by weight of an auxiliary micelle-forming non-soap surfactant selected from:

(b) (i) (i) anionic non-soap surfactants other than those defined under (a) above, ethoxylated nonionic surfactants having HLB values of at least 12, and amine oxides; or

(b) (i) (ii) fatty acid mono- and diethanolamides, and ethoxylated nonionic surfactants having HLB values lower than 12;

and/or

(b) (ii) 1 to 15% by weight of an additive which is a non-micelle-forming or weakly micelle-forming aliphatic, alicyclic, aromatic or araliphatic nonionic material having a melting point not exceeding 55°C and a dielectric constant of its liquid form of at least 2.2, selected from:

(b) (ii) (i) materials containing at least 4 carbon atoms, and containing a hydroxyl group positioned terminally or within 2 carbon atoms of the terminal position on a hydrocarbon chain

2

or on an aromatic or alicyclic ring, and optionally one or more further polar groups; containing not more than 6 aliphatic carbon atoms if the hydroxyl group is the only polar group present or if a second polar group is present and separated from the hydroxyl group by 2 or less carbon atoms; or containing not more than 12 aliphatic carbon atoms if a second polar group is present and separated from the hydroxyl group by 3 or more carbon atoms; or

(b) (ii) (ii) materials containing at least 7 carbon atoms, and containing at least one polar group positioned terminally or within 2 carbon atoms of the terminal position on a hydrocarbon chain or on an aromatic or alicyclic ring, and/or containing ethylenic unsaturation; the total amounts of (a) plus (b) being within the range of from 15 to 95% by weight;

and, optionally,

(c) from 1 to 45% by weight of a second additive which is an anionic or nonionic water-soluble non-micelle-forming or weakly micelle-forming material having a polar head group and optionally an aliphatic or araliphatic hydrocarbon chain containing at most 6 aliphatic carbon atoms;

component (c) being essential if neither an auxiliary surfactant (b) (i) (i) nor an additive (b) (ii) (i) is present; and

(d) water.

The first-mentioned surfactant (a) may, if desired, be an alpha-sulphonated fatty acid salt or a fatty acid methyl ester sulphonate. Such compositions form no part of the present invention.

DISCLOSURE OF THE INVENTION

The detergent gels of the invention are characterised by being wholly or predominantly in hexagonal liquid crystal form. This crystal form, also known as "middle" phase, may be recognised by various microscopic techniques, of which X-ray diffraction is the most definitive:

hexagonal phase compositions give rise to a characteristic X-ray diffraction pattern unique to this liquid crystal form. The ratio between the Bragg spacings in the X-ray pattern is:

$$\frac{1}{\sqrt{3}} : \frac{1}{\sqrt{4}} : \frac{1}{\sqrt{7}}$$

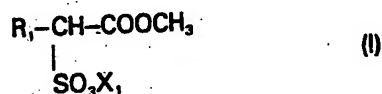
see, for example, "Liquid Crystals and Plastic Crystals", edited by G W Gray and P A Winsor (Ellis Horwood Limited, 1974), volume 2, chapter 4, page 88.

Of the three liquid crystal forms—lamellar, hexagonal and cubic—hexagonal phase is intermediate in rigidity, but rigidity or viscosity is not an infallible means of distinguishing hexagonal phase gels from other gels because, for example, softer lamellar phase gels can be thickened with polymers or electrolytes to produce products of comparable viscosity. The products of the invention are inherently stiff gels, and require no thickening agent. Preferred embodiments are transparent or translucent, and are sufficiently attractive in appearance for packing in transparent containers.

The surfactant component(a)

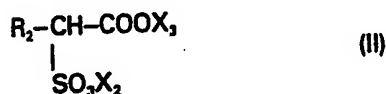
The surfactant component (a) is selected from alpha-sulphonated fatty acid salts, fatty acid methyl ester sulphonates, and mixtures of the two.

Fatty acid methyl ester sulphonates (FAES) are materials of the general formula I



wherein R_1 is an alkyl group having from 6 to 20 carbon atoms and X_1 is a solubilising cation, preferably alkali metal, ammonium, or substituted ammonium. Fatty acid methyl ester sulphonates may be substantially of a single chain length, or may consist of mixtures of materials of different chain length, as in coconut FAES (C_8-C_{18} , with R_1 as C_{10} and C_{12} predominating).

Alpha-sulphonated fatty acid salts (SFAS) are materials of the general formula II



wherein R_2 is an alkyl group having from 6 to 20 carbon atoms, X_2 is a solubilising cation, and X_1 is H or a solubilising cation (the same as or different from X_2), depending on pH.

With FAES, the choice of the solubilising cation is not critical. SFAS, however, must be wholly

different, selected from alkyl, alkenyl, aryl, alkaryl, aralkyl, hydroxyalkyl and alkoxyalkyl groups. Examples include mono-, di- and trialkylamines, for example, ethylamines, and mono- di- and trialkanolamines, for example, ethanolamines.

- It is already known from the aforementioned US 2 580 713 that the range of concentrations over which stable hexagonal phase gels based on lauryl sulphate can be obtained is larger if the counter-cation is non-metallic, such as triethanolamine, than if it is metallic, such as sodium, and a similar effect has been observed with the gels of the present invention. For alpha-sulpho fatty acid salts alone, stable hexagonal phase gels cannot be obtained at all from the monosodium, disodium or monoammonium salts, while the triethanolamine salts will give stable gels. Fatty acid methyl ester sulphonates and mixtures are more amenable and their sodium salts will form gels spontaneously at ambient temperature, but the range of concentrations over which stable gels can be obtained appears to be larger for the ammonium or substituted ammonium salts.

Especially preferred are compositions in which SFAS is wholly or predominantly in triethanolamine (TEA) salt form. Compositions in which SFAS is in 100% TEA salt form are of particular interest.

With FAES-based compositions, on the other hand, there is no preference for TEA salt-based compositions over sodium salt-based compositions.

The auxiliary surfactant (b)

- If desired, an auxiliary surfactant (b) may be present. This may be a fatty acid soap, or a defined non-soap anionic surfactant.

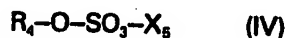
Fatty acid soaps are materials of the general formula III:



- wherein R_3 is an alkyl or alkenyl group having from 9 to 20 carbon atoms and X_4 is a solubilising cation, for example, alkali metal, ammonium, substituted ammonium or magnesium.

The auxiliary surfactant may alternatively or additionally comprise a non-ethoxylated micelle-forming non-soap anionic surfactant which is "primary" in nature, that is to say, the anionic head group is positioned terminally or next to terminally on the C_{10} - C_{20} aliphatic or araliphatic hydrocarbon chain. Examples of such surfactants include those listed below.

(i) Primary alkyl sulphates of the general formula IV



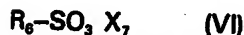
- wherein R_4 is an alkyl group having from 10 to 20 carbon atoms and X_5 is a solubilising cation. Primary alkyl sulphates may be substantially of a single chain length, as in dodecyl sulphate; or they may consist of mixtures of materials of different chain lengths, as in coconut alkyl sulphate (C_{10} - C_{14} , with C_{12} and C_{14} predominating).

(ii) Alpha-olefin sulphonates of the general formula V:



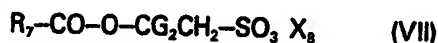
- wherein R_5 is an alpha, beta-unsaturated C_{10} - C_{20} alkenyl group and X_6 is a solubilising cation.

(iii) Primary alkane sulphonates of the general formula VI:



wherein R_6 is a primary C_{10} - C_{20} alkyl group and X_7 is a solubilising cation.

(iv) Alkyl or alkenyl isethionates of the general formula VII:



wherein R_7 is a C_7 - C_{20} alkyl or alkenyl group and X_8 is a solubilising cation.

Amounts of surfactant present

In the compositions of the invention, the total level of surfactant present is from 25 to 95% by weight. If no auxiliary surfactant (b) is present, the total level of FAES and/or SFAS must be from 25 to 95% by weight, while lower levels of these ingredients (5-25%) are possible when an auxiliary surfactant (b) is present.

- Preferably the total surfactant level is within the range of from 50 to 85% by weight. The total level of FAES and/or SFAS present, whether or not an auxiliary surfactant is also present, is preferably from 40 to 85% by weight.

The preferred range for FAES is from 10 to 65% by weight. When FAES is the sole surfactant, the preferred range is from 50 to 65% by weight. The most preferred range will of course be lower for the sodium salt than for the higher-molecular-weight triethanolamine salt.

The preferred range for SFAS is from 5 to 85% by weight. When SFAS is the sole surfactant, wholly or predominantly in the form of the salt of a substituted ammonium cation, the preferred range is from 60 to 85% by weight.

When FAES and SFAS are used together, the weight ratio of FAES to SFAS may conveniently range from 15:1 to 1:15, preferably from 11:1 to 1:6.

When an auxiliary surfactant (b) is present, lower amounts of FAES and/or SFAS may generally be used. The amount of auxiliary surfactant (b) is from 1 to 75% by weight, preferably from 5 to 30% by weight and especially from 15 to 25% by weight. The weight ratio of FAES and/or SFAS to auxiliary surfactant may conveniently lie in the range of from 15:1 to 1:15, preferably from 5:1 to 1:5.

The additive (c)

The amount of electrolyte that can be tolerated in compositions of the invention may be increased by the addition of an additive (c) as defined previously. This material is described in detail in the aforementioned GB 2 155 031A (Unilever). It is a water-soluble non-micelle-forming or weakly micelle-forming material capable of driving an aqueous surfactant system from lamellar phase into hexagonal phase, or of increasing the region of stable hexagonal phase available with a particular composition. The mechanism of action of the additive is not clearly understood; it is possible that it acts so as to increase micelle or liquid crystal curvature, but the scope of the invention is not to be limited by this hypothesis. Empirically it has been observed that some materials useful as hydrotropes in light-duty liquid detergent compositions may behave as additives. These are generally molecules containing a large polar group and, optionally, a small hydrophobic group, such as an aliphatic or araliphatic chain containing not more than 6, preferably 4 or less, aliphatic carbon atoms. The larger the polar head group, the larger the hydrophobe that can be tolerated.

The polar group of the additive may carry an ionic charge, but if so this must be of the same polarity as that of the surfactant or surfactants, that is to say, anionic. Examples of anionic additives are the lower aryl or alkylaryl sulphonates, such as toluene and xylene sulphonates.

Alternatively the additive may be a highly polar but uncharged material.

A preferred type of uncharged additive is typified by the lower amides, containing the

-CON- group.

Common features of this type appear to be an ability to raise the dielectric constant of water combined with a structure-breaking effect on water. The preferred material, which is both cheap and environmentally unobjectionable, is urea. Short-chain urea homologues and analogues, for example, methyl and ethyl ureas, thiourea, formamide and acetamide, are possible alternatives, but these are of less interest than urea itself in view of various drawbacks such as cost, toxicity or simply a lesser effectiveness as an additive.

If the additive (c) is urea, a buffering agent is advantageously present in order to minimise hydrolysis, especially alkaline hydrolysis, of the urea. A suitable buffer is boric acid, preferably used in an amount of less than 3% by weight, more preferably from 1 to 2% by weight. In systems where TEA is present as a cation this may, however, be unnecessary because of the buffering capability of TEA. Its electrolyte tolerance also allows larger quantities of electrolytic ingredients, such as sodium tripolyphosphate and other builders, to be included than when the surfactants are wholly in the form of the salts of non-buffering cations such as sodium.

The additive (c) is advantageously present at a level of from 1 to 45% by weight, preferably from 5 to 35% by weight, more preferably from 5 to 15% by weight. The higher the electrolyte level, the more additive will be required.

The water (d)

In all embodiments of the invention, water is an essential constituent. The relative proportions of any particular system of surfactant and water that will give stable hexagonal phase or predominantly hexagonal phase gels can be ascertained by experiment, and a phase diagram constructed. Samples at various ratios are prepared by mixing, and the phases present can be recognised without difficulty by visual appearance, gross flow properties, appearance in polarised light, and texture observed in a polarising microscope.

Optional additional ingredients

Unbuilt compositions of the invention may consist substantially or wholly of surfactant component (a), optional additive, and water, plus the usual minor ingredients such as colour, perfume, germicides and preservatives. Such unbuilt compositions are useful for light-duty applications, for

ingredients such as builders, bleaches, fluorescers, photobleaches, enzymes, antiredeposition agents, deoperfumes and germicides. Water-soluble organic or inorganic builders, for example, phosphates, citrates or nitrilotriacetates, may be incorporated in the composition of the invention but, as indicated previously, care must be taken that the electrolyte level does not rise to such an extent that the hexagonal phase gel is destabilised. As indicated previously, the amount of electrolyte builder that can be incorporated can be increased by also including an additive (c), preferably urea. The same considerations apply to the inclusion of other electrolytic ingredients, for example, sodium hypochlorite or sodium sulphite bleaches.

Alternatively, water-insoluble inorganic builders such as zeolite may be suspended in the gels of the invention. Equally, other useful insoluble materials such as abrasives or peroxyacid bleaches may be present as suspended solids. An especially preferred bleach material is 1, 12-diperoxidodecanedioic acid, as described in EP 160 542A (Unilever).

EXAMPLES

The following non-limiting Examples, in which percentages and ratios are by weight, illustrate the invention.

Compositions were prepared by mixing from the ingredients shown in the various Tables; the numbered compositions are in accordance with the invention while the lettered ones are comparative. The coconut FAES contained approximately 5% SFAS as an impurity.

Examples 1 to 4 are compositions containing coconut FAES (sodium or ammonium salt) and water only. Stable gels containing 55–60% by weight of either salt could be prepared, as shown below:

Example	Coconut FAES (Cation)	Water	Product
A	50 (Na)	50	Liquid
1	55 (Na)	45	Stable gel
2	55 (NH ₄)	45	Stable gel
3	60 (Na)	40	Stable gel
4	60 (NH ₄)	40	Stable gel
B	80 (Na)	20	Lamellar phase

Examples 5 to 9 are compositions containing coconut SFAS (mono TEA salt) and water only: TEA denotes the triethanolamine cation. Stable gels containing 60–75% by weight of the surfactant could be prepared, as shown below:

Example	Coconut SFAS (monoTEA)	Water	Product
C	45	55	Liquid
D	50	50	Cubic
5	60	40	Stable gel
6	65	35	Stable gel
7	70	30	Stable gel
8	75	25	Stable gel
9	85	15	Stable gel

Comparative Examples E to J are compositions containing coconut SFAS (monosodium salt) and water only. It was not possible to prepare hexagonal phase gels using this combination of ingredients.

Example	Coconut SFAS (MonoNa)	Water	Product
5 E	20	80	Solution plus crystals
F	30	70	Mass of crystals
G	40	60	Mass of crystals
H	50	50	Mass of crystals
10 J	60	40	Mass of crystals

Examples 10 and 11 are compositions containing both FAES and SFAS.

Example	Coconut FAES (cation)	Coconut SFAS (cation)	Water	Product
10	55 (Na)	5 (monoNa)	40	Stable gel
20 K	50 (Na)	10 (monoNa)	40	Liquid
11	10 (TEA)	60 (monoTEA)	30	Stable gel
L	10 (TEA)	55 (monoTEA)	35	Cubic

25 Example 12
A stable gel containing:

	%
Coconut FAES (Na salt)	40
Coconut alkyl sulphate (Na salt)	20
Water	40

35 was prepared

40 Example 13
A stable gel containing:

	%
45 Coconut FAES (Na salt)	40
Coconut soap (Na salt)	20
Water	40

50 was prepared.

Examples 14 to 16 are FAES-based compositions built with sodium tripolyphosphate. It will be noted that the FAES is able to tolerate a relatively high electrolyte level, but the range of possible formulations is extended by the inclusion of urea.

5 Example	Coconut FAES (sodium)	Builder	Urea	Water	Product	5
M	50	10	—	40	Flowable liquid	
14	50	10	10	30	Gel plus builder crystals	
10 15	55	10	—	35	" " "	10
16	55	10	10	25	" " "	

15 *Examples 17 and 18 are compositions based on coconut FAES containing sodium hypochlorite (used to bleach the FAES, and possibly partially decomposed to sodium chloride): this is a common impurity in FAES. The inclusion of urea allowed stable gels to be prepared despite the high electrolyte level.* 15

20 Example	Coconut FAES (sodium)	Sodium hypochlorite	Urea	Water	Product	20
25 N	65	2	—	33	Lamellar phase	25
17	65	2	10	23	Stable gel	
P	65	3	—	32	Lamellar phase	
18	65	3	10	22	Stable gel	

30 *Example 19*
A stable gel containing

35		%	35
	Coconut FAES (Na salt)	50	
	Sodium sulphite	5	
40	Urea	10	40
	Water	35	

45 was prepared. The sodium sulphite functions as a mild bleaching agent, as described in GB 1 417 840 (Unilever) and British Patent Application No 86 06145 (Unilever). As with Examples 14 to 18, the inclusion of urea allowed a stable gel with a high electrolyte level to be prepared. 45

Examples 20 to 28

50 Stable FAES-based gels suitable for fabric washing were prepared as shown in the Table, the base gel being that of Example 3 (60% sodium coconut FAES). The fabric washing ingredients, used separately and in combination, were as follows: 50

Antiredeposition agent: sodium carboxymethyl cellulose, ex Courtaulds (62% active matter)

Proteolytic enzyme: alcalase solution ex Novo, activity 1600 glycine units/mg

55 Fluorescer: Tinopal (Trade Mark) CBS-X ex Ciba-Geigy, which is 4,4'-di(2-sulphostyryl) diphenyl, disodium salt. 55

Photobleach: aluminium phthalocyanine sulphonate ex Ciba-Geigy

Germicide: formalin (37%)

Deoperfume: ex PPF International

Buffer: citric acid

60 Solid peroxy acid bleach: 1,12-diperoxydodecanedioic acid (DPDDA) granules ex Degussa (12% available oxygen, produced from DPDDA granules ex Degussa slurried in water and filtered to remove water-soluble inorganic salts). 60

Examples 20 to 28 were stable translucent hexagonal phase gels.

The dishwashing performances of two unbuilt gels according to the invention were evaluated using a standardised test procedure in which soiled plates were washed to a foam collapse end point. Each plate was pre-soiled with 5 g of a standard cooking oil/starch/fatty acid emulsion in water, and the washing solution in each case consisted of 5 g of product dissolved in 5 litres of water (4° French hardness) at 45°C, that is to say, a whole product concentration of 1 g/litre. The gels tested were those of Examples 3 and 11. The results were as follows:

	3	11
Coconut FAES (Na salt)	60	—
Coconut FAES (TEA salt)	—	10
Coconut SFAS (monoTEA salt)	—	60
Water	40	30
Number of plates washed	18	15

20 21 22 23 24 25 26 27 28

[illegible]

CLAIMS

1. An aqueous detergent composition comprising a gel wholly or predominantly in hexagonal liquid crystal form, wherein the gel comprises:
 - 5 (a) from 5 to 95% by weight of a fatty acid methyl ester sulphonate and/or an alpha sulpho fatty acid salt, 5
 - (b) optionally from 1 to 75% by weight of an auxiliary surfactant selected from fatty acid soaps; non-ethoxylated micelle-forming non-soap surfactants having an anionic head group and an aliphatic or araliphatic hydrocarbon chain containing from 10 to 20 aliphatic
 - 10 carbon atoms, the anionic head group being positioned terminally or next to terminally in the hydrocarbon chain, and mixtures thereof; 10
 - the total amount of components (a) and (b) being from 25 to 95% by weight,
 - (c) optionally 1 to 45% by weight of an additive which is an anionic or nonionic water-soluble non-micelle-forming or weakly micelle-forming material having a polar head group and optionally
 - 15 an aliphatic or araliphatic hydrocarbon chain containing at most 6 carbon atoms, and 15
 - (d) water,
 - with the proviso that if component (a) consists wholly of an alpha-sulpho fatty acid salt, the alpha-sulpho fatty acid is wholly or predominantly in substituted ammonium salt form.
2. A composition as claimed in claim 1, wherein the alpha-sulpho fatty acid is wholly or
- 20 predominantly in triethanolamine salt form. 20
3. A composition as claimed in claim 1 or claim 2, which comprises from 50 to 85% by weight, in total, of surfactants (a) and (b).
4. A composition as claimed in any of claims 1 to 3, which comprises from 40 to 85% by weight of a fatty acid methyl ester sulphonate and/or alpha-sulpho fatty acid salt.
- 25 5. A composition as claimed in claim 4, which comprises from 10 to 65% by weight of a fatty acid methyl ester sulphonate. 25
6. A composition as claimed in claim 4, which comprises from 5 to 85% by weight of an alpha-sulpho fatty acid salt.
7. A composition as claimed in any one of claims 1 to 6, which comprises from 5 to 30%
- 30 by weight of an auxiliary surfactant (b). 30
8. A composition as claimed in any one of claims 1 to 7, comprising from 5 to 35% by weight of urea as an additive (c).
9. An aqueous detergent composition comprising a gel wholly or predominantly in hexagonal liquid crystal form, wherein the gel comprises:
 - 35 (a) from 15 to 85% by weight of a fatty acid methyl ester sulphonate and/or an alpha-sulpho fatty acid salt, 35
 - (c) optionally 1 to 45% by weight of an additive which is an anionic or nonionic water-soluble non-micelle-forming or weakly micelle-forming material having a polar head group and optionally an aliphatic or araliphatic hydrocarbon chain containing at most 6 carbon atoms, and
 - 40 (d) water, 40
 - within the proviso that component (a) cannot consist wholly of an alpha-sulpho fatty acid salt in sodium salt form.
10. A composition as claimed in claim 1, substantially as described in any one of the Examples 1 to 28 herein.

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